Search for Scintillation in Doped Lead Fluoride Crystals

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***Abstract*—An effort has been made to introduce scintillation light in lead fluoride crystals by selective doping. It was found that some rare earth ions doped in the crystal may serve as luminescence centers. The photo- and X- luminescence spectrum, the decay time constant and the light output of these doped samples are presented. Because of the slow decay time these doped lead fluoride samples fall short to be used to build a homogeneous hadronic calorimeter with dual readout for future high energy physics experiments. Researches will continue to introduce scintillation in lead fluoride.**

I. Introduction

INORGANIC crystal scintillators have played an important role in the construction of total absorption electromagnetic calorimeter (ECAL) for high energy physics experiments. Crystals have recently also been proposed to construct a homogeneous calorimeter, including both electromagnetic and hadronic parts [1] for detectors at the international linear collider. This homogeneous hadronic calorimeter (HHCAL) detector concept removes the traditional boundary between the ECAL and the HCAL, so eliminates the effect of dead materials in the middle of the hadronic shower development. It may also takes advantage of the recently implemented dual readout approach to achieve good energy resolution for hadronic jets by measuring both Čerenkov and scintillation light [2]. Because of the un-precedent volume (70 to 100 m^3) foreseen for such calorimeter [1], the crystal material must be dense (to reduce the volume), UV transparent (to effective collecting the Čerenkov light) and allows a clear discrimination between the Čerenkov and scintillation light.

Cubic lead fluoride (PbF_2) has a high density of 7.77 g/cm^3 and a short radiation length (X_0 = 0.93 cm) and a short nuclear interaction length (λ_I = 21 cm). It has good UV transparency down to 250 nm. Its low melting point (824°C) and low material cost (1/3 of BGO) makes it potentially a cost-effective material for the HHCAL detector concept. Large size (20 cm long) PbF_2 crystals are available in the market. They were used as a Čerenkov material to construct the ECAL for the A4 experiment at the MAinzer MIcrotron (MAMI) facility in Mainz. Excellent energy resolution was reported [3].

As a Čerenkov material PbF_2 has been studied in details [4]. The only issue for the HHCAL application is that it is not a scintillator. Effort was made to introduce scintillation light into PbF_2 through phase transition (cubic to orthorhomic) during growth. Positive result was reported by Klassen *et al.* [5], but was not confirmed by Derenzo *et al.* [6]. Observation of fast photo- and x- luminescence in Gd doped PbF_2 crystals was first reported by Shen *et al.* of SIC [7], and was later confirmed by Woody *et al.* in a beam test at AGS [8]. In this work lead fluoride samples doped with various rare earth elements were grown by modified Bridgman method. Photo- and X- luminescence spectrum, decay kinetics and γ-ray excited anode current and pulse height spectrum were measured.

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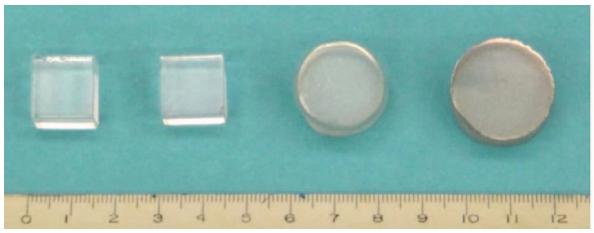

Fig. 1. A photo showing lead fluoride crystal samples doped with various rare earth elements from SIC (left two cubes) and Scintibow (right two cylinders).

A total of 116 PbF_2 samples doped with various rare earth elements were grown by modified Bridgman method at Shanghai Institute of Ceramics (SIC) and Shanghai Scintibow Crystal Co., LTD. While classical platinum crucible was used at SIC for the crystal growth, graphite crucible was used at Scintibow. The SIC samples are cubes of 1.5 radiation length with six faces polished as shown in the left two samples in Fig.1. Most Scintibow samples are cylinders with two end faces polished and have a dimension of ϕ 22 $\times$ 15 mm as shown in the right two samples in Fig.1. Crystals grown in graphite crucible are less transparent that in platinum crucible.

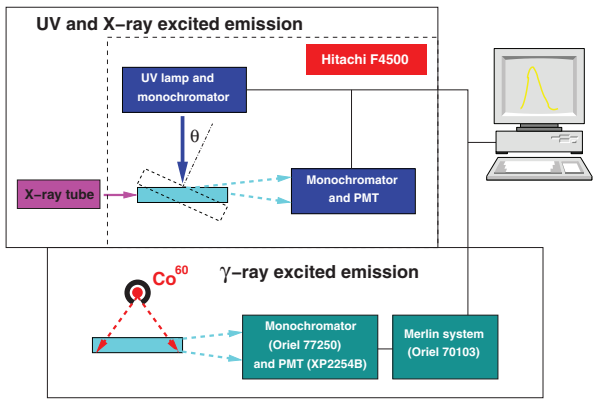

Fig. 2. The setup used for the photo- and x- luminescence measurement.

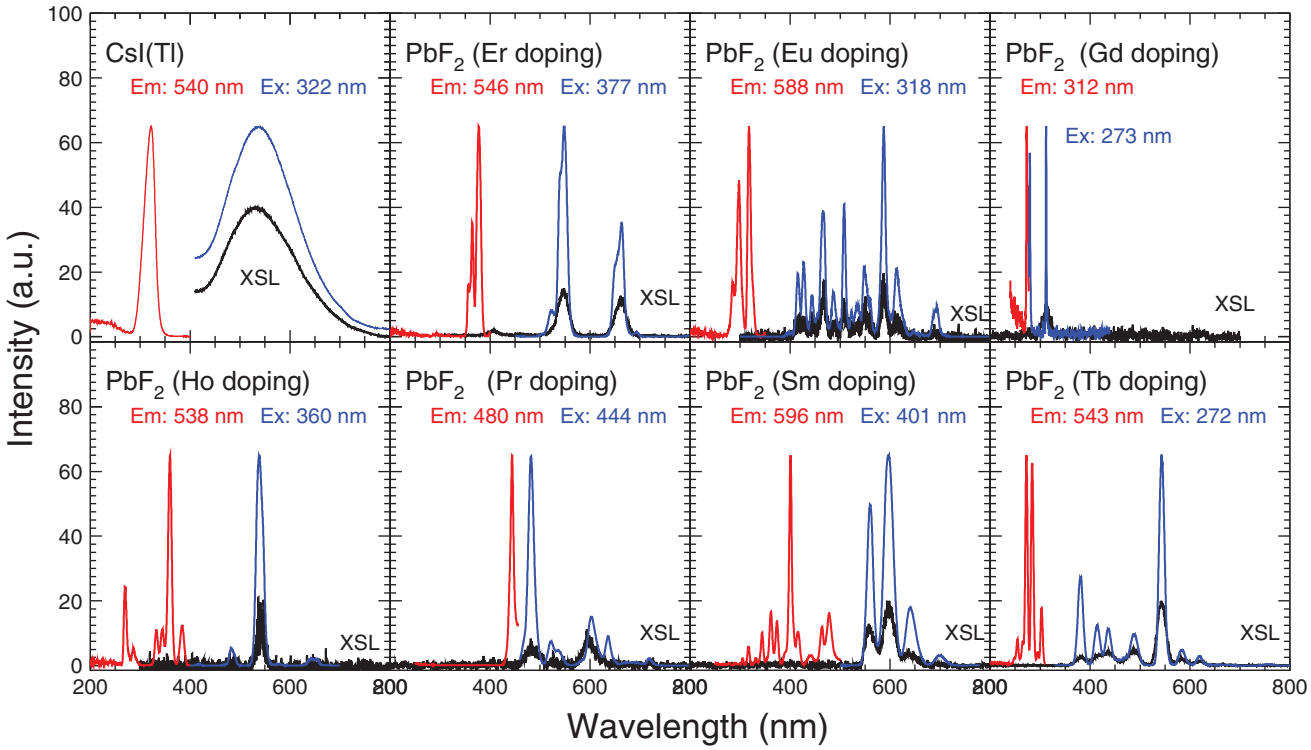

Fig. 3. The excitation (red) and Photo- (blue) and X- (black) luminescence spectra are shown as a function of wavelength for the PbF_2 samples doped with Er, Eu, Gd, Ho, Pr, Sm and Tb as well as a reference CsI(Tl) sample.

II. Photo- and X- Luminescence Spectrum

Photo- and X-ray luminescence spectrum was measured by a Hitachi F-4500 fluorescence spectrophotometer. Fig. 2 shows the set-up used for this measurement. For the X-ray luminescence measurement an AMTPEK E3-T X-ray tube was run at 25 kV and 50 μA. Fig. 3 shows excitation (red) photo-luminescence (blue) and X-luminescence (black) spectra for the PbF_2 samples doped with Er, Eu, Gd, Ho, Pr, Sm and Tb as well as a reference CsI(Tl) sample. Although spectroscopic resolutions are different, the observed photo- and X- luminescence spectra are consistent for all samples.

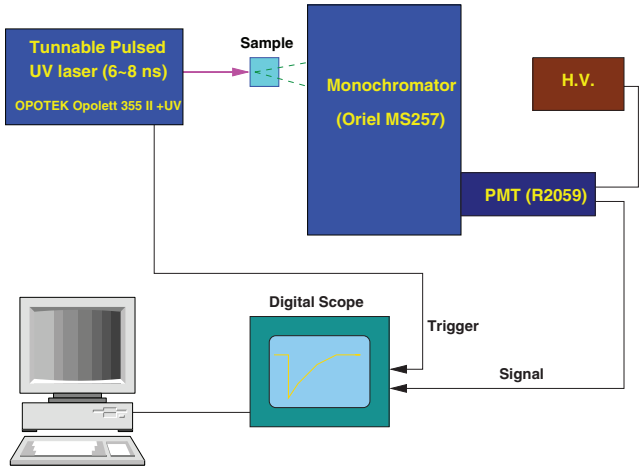

Fig. 4. The setup used for the photo-luminescence pulse shape and the decay time measurement.

The photo- and x- luminescence spectra observed in the Gd doped PbF_2 samples are consistent with previous publications by Shen and Woody [7], [8].

III. Photo-luminescence Decay Time Constant

The photo-luminescence decay time constant of these doped PbF_2 samples was measured by using a pulsed laser as the excitation source. Fig. 4 shows the set-up used in this measurement. The UV light pulses of 6 – 8 ns width from an *OPOTEK Opolette 355 II + UV* tunable laser was used as the excitation source. The photo-luminescence light from these samples went first through an Oriel MS257 monochromator then to a Hamamatsu R2059 PMT. The luminescence pulse shape observed by the PMT was recorded by an Agilent 6052A digital scope. The decay time constants were determined by an exponential fit to the pulse shape. Fig. 5 shows the photo-luminescence pulse shape (blue), the corresponding exponential fit (red dots) and decay time constant for the PbF_2 samples doped with Er, Ho, Eu, Sm and Tb as well as a reference CsI(Tl) sample. Table I summarizes the decay time constants for samples doped with rare earth, which were found at a millisecond scale. This time constant is too long to be useful for high energy physics experiments.

TABLE I
Decay Time Constant for Doped Lead Fluoride Crystals

Dopant	Er	Eu	Ho	Sm	Tb
Decay time constant (ms)	1.5	8.5	1.3	7.0	5.0

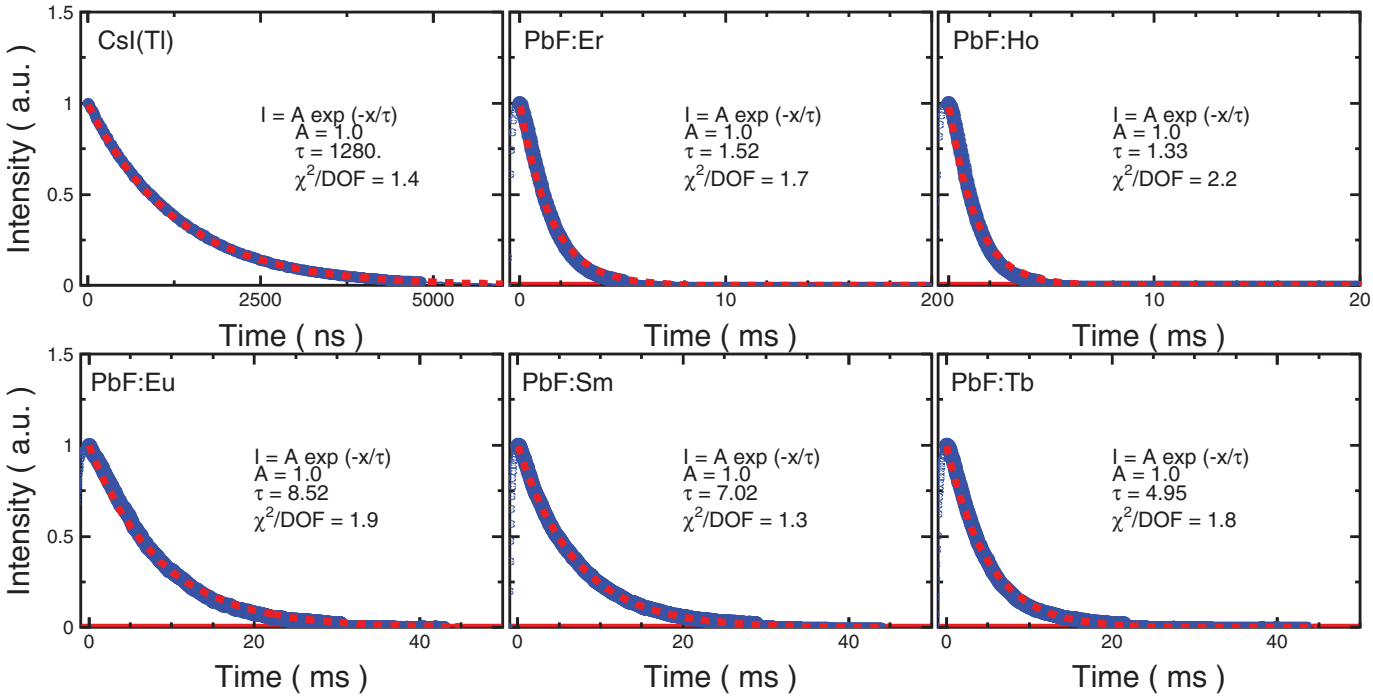

Fig. 5. The photo-luminescence pulse shape (blue), corresponding fit to an exponential (red dots) and the decay time constant are shown for the PbF_2 samples doped with Er, Ho, Eu, Sm and Tb as well as a reference CsI(Tl) sample.

IV. γ-ray Induced Anode Photo-Current

Fig. 6 shows the setup used for the anode current measurement. A ^{137}Cs source was used to excite the samples. A Hamamatsu R2059 PMT was used to measure the anode photo-current. The bias voltage of the PMT was fixed at -2,000 V, and the distance between the source and the samples was fixed at 2 cm. The PMT anode current was measured by using a digital multi-meter.

Fig. 7 and Fig. 8 show the anode photo-current measured respectively for all PbF_2 samples (black and blue dots) and a un-doped PbF_2 sample (red). The corresponding anode photo-current of a reference PWO sample with light output of 20 p.e./MeV is also shown in Fig. 8. The anode photo-current was found to be 42 nA and 240 nA respectively for the un-doped PbF_2 and the PWO sample. A total of six doped PbF_2 samples showed an anode photo-current of larger than 50

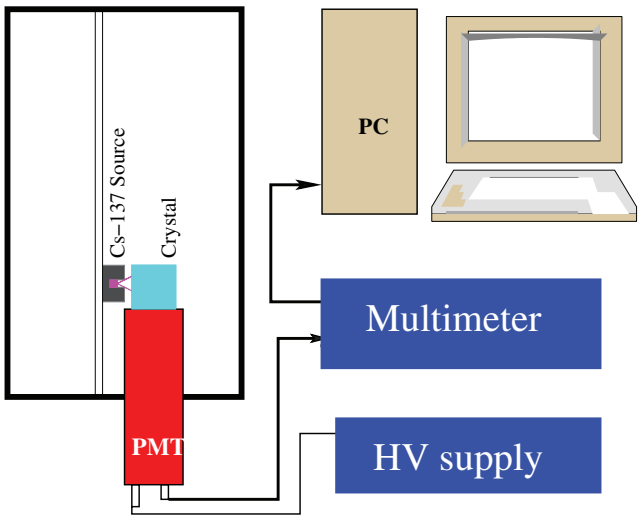

Fig. 6. The setup used to measure the γ-ray induced anode photo-current for PbF_2 samples. The distance between source and samples was fixed at 2 cm.

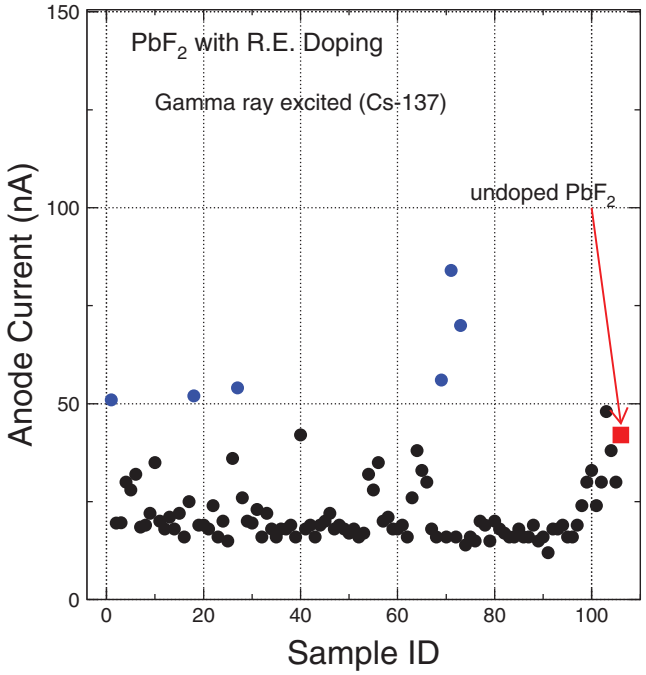

Fig. 7. The PMT anode photo-current measured for all PbF_2 samples (black and blue dots).

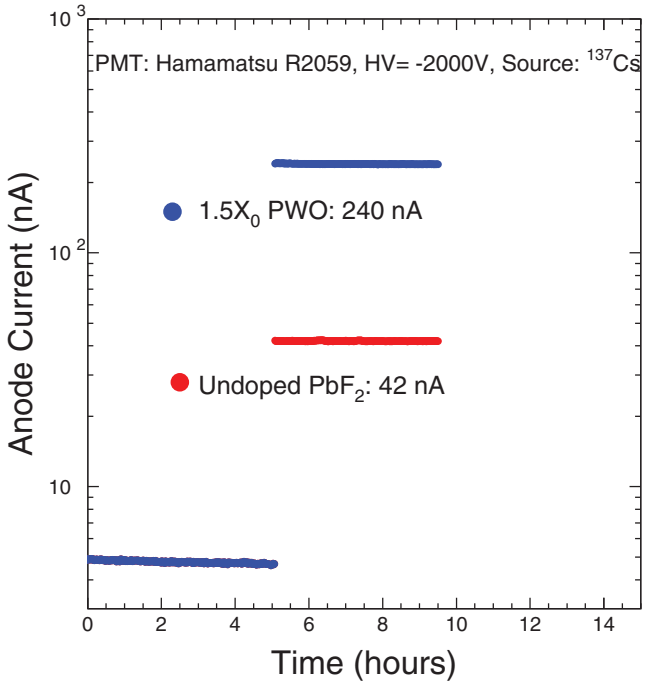

Fig. 8. The PMT anode current measured for a un-doped PbF_2 sample (red) and a reference small PWO sample (blue) with light output of 20 p.e./MeV

nA, which are marked as the blue dots in Fig. 7. Table II lists their ID, dimension and dopant. The numerical result of this measurement indicates that the light output of these doped PbF_2 samples are less than 3 p.e./MeV. We also note that all these samples are doped with Eu, and were grown at Scintibow.

TABLE II
THE PMT ANODE CURRENT FOR DOPED PbF_2 SAMPLES

ID	Anode Current (nA)	Size (mm)	Doping
Scintibow-1	51	18 × 12 × 10	Eu
Scintibow-18	52	ϕ 22 × 15	Eu / Gd
Scintibow-27	53	ϕ 20 × 15	Eu / Tb
Scintibow-B19	56	ϕ 20 × 15	Eu / Tb / Na
Scintibow-B21	83	ϕ 22 × 15	Eu / Bi / Na
Scintibow-B23	73	ϕ 20 × 15	Eu / Bi / Na

V. ^{137}Cs γ-ray Exited Pulse Height Spectrum

^{137}Cs γ-ray exited pulse height spectrum was measured for doped PbF_2 samples listed in table II. A Hamamatsu R2059 PMT was used as the readout device with an integration gate of 1 μs. Fig. 9 and Fig. 10 show the pulse height spectra for samples Scintibow-1 and Scintibow-B21 respectively. Also shown in these figures is the pulse height spectrum for a un-doped PbF_2 sample. There is very little difference between spectra obtained from the doped and the un-doped PbF_2 samples, indicating that the amount of the luminescence light in the 1 μs gate is too weak to show a peak.

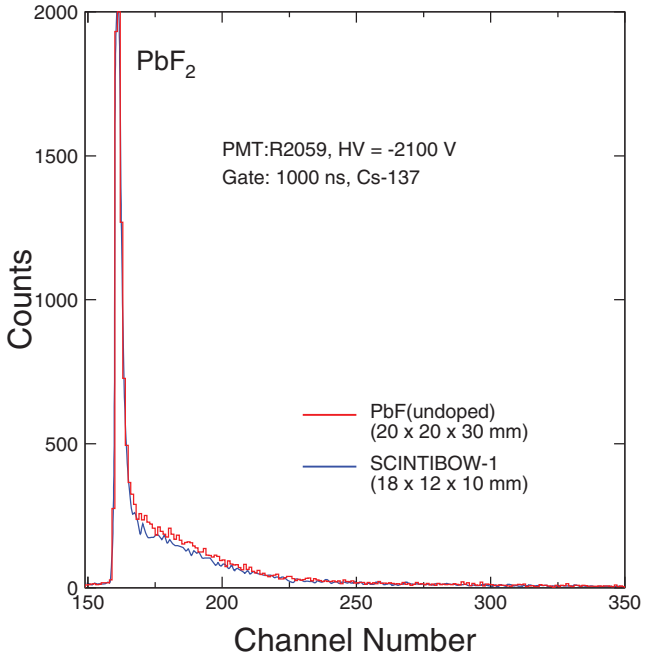

Fig. 9. ^{137}Cs γ-ray exited pulse height spectrum for doped PbF_2 samples Scintibow-1.

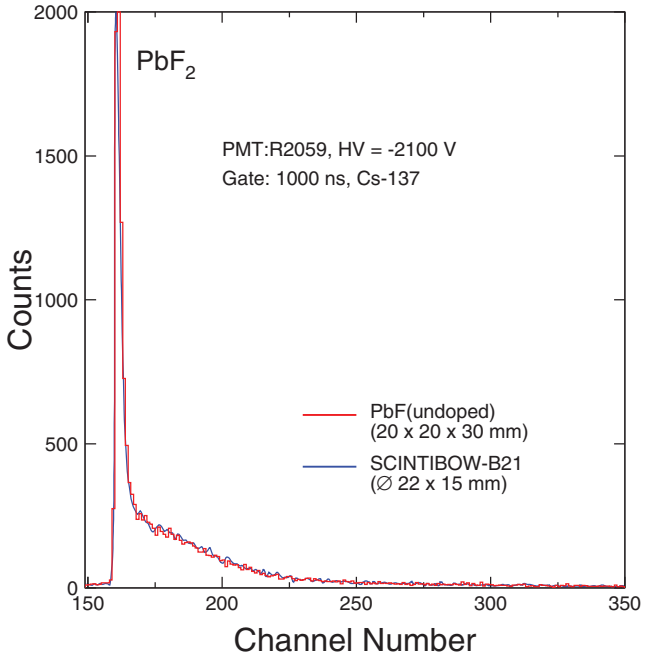

Fig. 10. ^{137}Cs γ-ray exited pulse height spectrum for doped PbF_2 samples Scintibow-B21.

VI. Summary

A search for scintillation was carried out in a set of doped lead fluoride crystal samples. Consistent photo- and x-ray luminescence spectra were found in PbF_2 samples doped with Er, Eu, Gd, Ho, Pr, Sm and Tb. The decay time of these doped samples was found to be at a millisecond scale as expected from the f-f transition of these rare earth elements. While some doped samples show γ-ray induced anode photo-current larger then the un-doped sample the numerical result indicates that the scintillation light, if any, is less than 3 p.e./MeV measured

by a PMT with bi-alkali photo-cathode. Their ^{137}Cs γ-ray excited pulse height spectra measured with 1μs integration gate were found identical to un-doped sample, confirming that their luminescence in the 1 μs gate is too weak to show a peak. Investigation will continue to search for scintillation in doped lead fluoride for the HHCAL detector concept.

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